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Methane consumption in aerated soils of the temperate zone

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ABSTRACT

Quasi-continuous observations of the methane concentration in aerated soils of the temperate zone are presented. The flux of methane into the soil is calculated from the concentration gradient at the soil surface. Depending on the permeability of methane in soil air at an individual site, yearly mean uptake rates between 0.09 and 1.3 g CH_4 m^{-2} yr^{-1} have been observed in temperate forest soils. The methane flux from the atmosphere into the soil shows only a weak seasonality with an about 50% higher uptake rate in the summer than in the winter months. This indicates that the methane flux into the soil is mainly controlled by the transport resistance in the soil rather than by the potential microbial decomposition rate.

1. Introduction

Oxidation of methane does occur in aerated soils by methylotrophic bacteria (Ehhalt, 1973; Haber et al., 1983, and references therein). Flux measurements in tropical (Keller et al., 1983, 1986) and subtropical soils (Seiler et al., 1984) have indicated that the total methane destruction rate in these regions plays a rather unimportant role, and may attribute less than 5% to the total global methane sink, i.e., photochemical destruction by OH radical. Flux measurements in the temperate zone and at different kinds of soil are, however, still very sparse.

The methane flux at the soil surface and the methane concentration in the soil air depend on both, the destruction rate by microbial decomposition, and the gas transfer in the soil air by molecular diffusion. In this study, gas transport in the soil is determined using the radioactive noble gas ^{222}Radon as a transport tracer (Dörr and Münnich, this issue). In this way we are able to distinguish between pure transport effects and effects by the microbial activity itself. ^{222}Radon is produced by radioactive decay of ^{226}Radium which is distributed rather uniformly in the soil matrix. The concentration profile of ^{222}Radon and the ^{222}Radon flux at the soil surface are, therefore, only controlled by individual soil texture and thus provide a measure of gas transport parameters in the soil at an individual site. ^{222}Radon and methane transport in the liquid phase can be neglected due to their comparably small solubility and the low soil moisture content in the unsaturated zone. With measurements of the ^{222}Radon concentration profile and the ^{222}Radon flux at the soil surface in parallel to the methane concentration profile in the soil air it is possible to determine the methane flux across the soil/atmosphere interface, and the parameters controlling this flux.

Direct methane fluxes are measured by placing an inverted cup at the soil surface and observing the methane concentration change with time. They are found to yield unreliable flux data due to a considerable disturbance of the steady state concentration profile in the soil, respectively the concentration gradient at the soil surface (Dörr and Münnich, this issue). The method used here, taking low-volume soil air samples, does, however, only very slightly disturb the steady state conditions. Using the ^{222}Radon tracer method we have determined the methane flux in different kinds of soil in the surroundings of Heidelberg over a time period of about one year and spot measurements at selected sites all over Europe.

The results and a discussion on the relevance of this methane sink on the global methane budget are presented here.

2. Methods

2.1. Experimental

Soil air samples are taken with a gas tight membrane pump through a stainless steel tube (2 mm inner diameter). The air is sucked through 8 holes of 0.5 mm diameter each just behind the cone tip of the probe. For a sampled soil gas volume of ca. 250 ml and a soil porosity of 0.2, the maximum depth resolution is ± 7 cm (see cf. vertical error bars in Fig. 1). Samples are collected in bags made of polyethylene coated weldable aluminum foil (Kalle, Wiesbaden, FRG) with teflon stopcocks (Peter von Berg, Kirchseeon, FRG) normally used for medical applications. These bags have been tested for leakage and memory effects: for storage times of several months and a methane concentration range from zero to ambient air concentration (≈ 2 ppmv) no significant changes of the sample air concentration in the bags was observed. Methane concentration is determined by GC/FID (Sichromat 1, Siemens, Karlsruhe, FRG). We use an evacuated gas inlet system, a sample loop of 3 ml, and a 1.5 m $\frac{1}{4}''$ molecular sieve 5A column maintained at 70°C (Born, 1988). The total reproducibility (1σ) of a single concentration measurement in soil air is ± 8 ppbv, the detection limit of the GC system is 20 ppbv. Methane concentrations are given relative to the NBS (National Bureau of Standards, Washington DC, USA) standard reference material SRM no. 1658.

From 100 ml aliquots of the samples ^{222}Radon activity is determined in a slow pulse ionization chamber (Roether and Kromer, 1978). The reproducibility (1σ) of a single ^{222}Radon measurement is typically $\pm 5\%$. Direct flux measurements for ^{222}Radon are carried out by the inverted cup method: the fluxes are calculated from the concentration increase in a container placed at the soil surface after an approximate accumulation period of half an hour to one hour. Due to the large relaxation depth of 180 cm the concentration increase under the inverted cup does, contrary to methane, *not* significantly disturb the ^{222}Radon concentrahon profile.

2.2. Sampling locations

Soil air samples have been collected periodically every one to two weeks at five different forest sites near Heidelberg using permanently installed sampling devices. The characteristics of the individual sites are given in Table 1. In addition, solitary samples from other cultivated soils in Western Germany and from an undisturbed volcanic soil close to the clean air station Izana (Tenerife, Canary Islands (Schmitt et al., 1988)) have been taken.

3. Results and discussion

Three typical concentration profiles of methane in soil air are given in Fig. 1 (upper part). A steep concentration decrease with depth can be observed at all three locations. At about 60 cm depth methane concentration has already decreased to nearly zero which must be attributed to the occurrance of a sink of methane in these soils.

The observed concentration profiles can be explained using the following model assumptions: if the methane sink (distribution of microbes) Q is homogeneously distributed hori-

Table 1. *Characteristics of the periodically used sampling sites near Heidelberg. Soil pH and soil organic matter (C_{org} in weight %) are given as mean values over the first 50 cm*

Sampling site	Vegetation type	Soil type	pH	C_{org}
M1 Sandhausen	beech/spruce	sandy soil	7.7	2.0
M2 Sandhausen	beech/oak/maple	loamy soil	8.0	2.3
M3 Sandhausen	beech/spruce	sandy soil	3.5	5.0
M4 Nußloch	spruce	clay	5.0	3.5
M5 Nußloch	mixed deciduous	clay	5.0	4.0

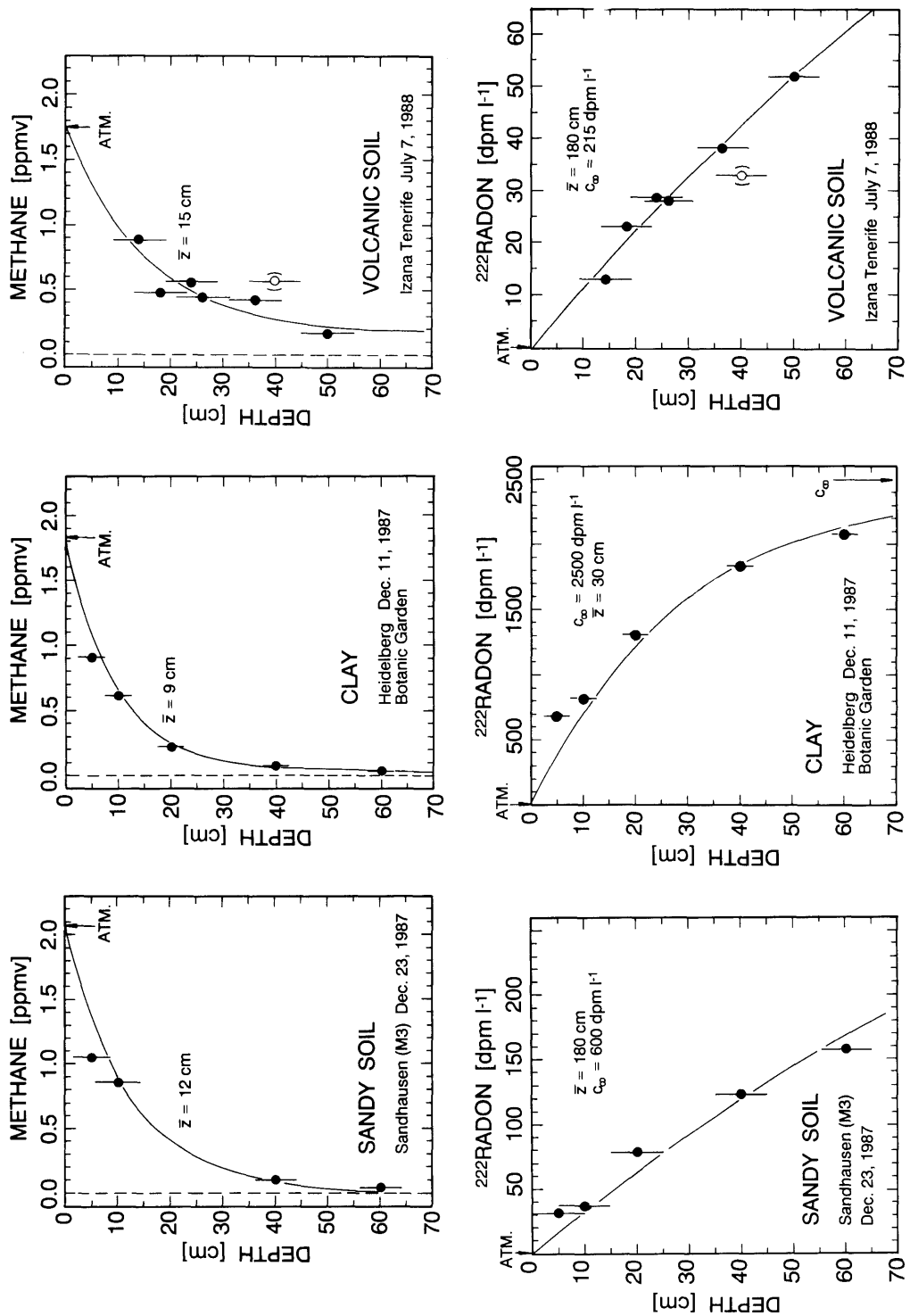

Fig. 1. Upper part: Concentration profiles of methane in the soil air at three different locations with different types of soil. Lower part: Corresponding ^{222}Radon profiles in the soil at the same locations.

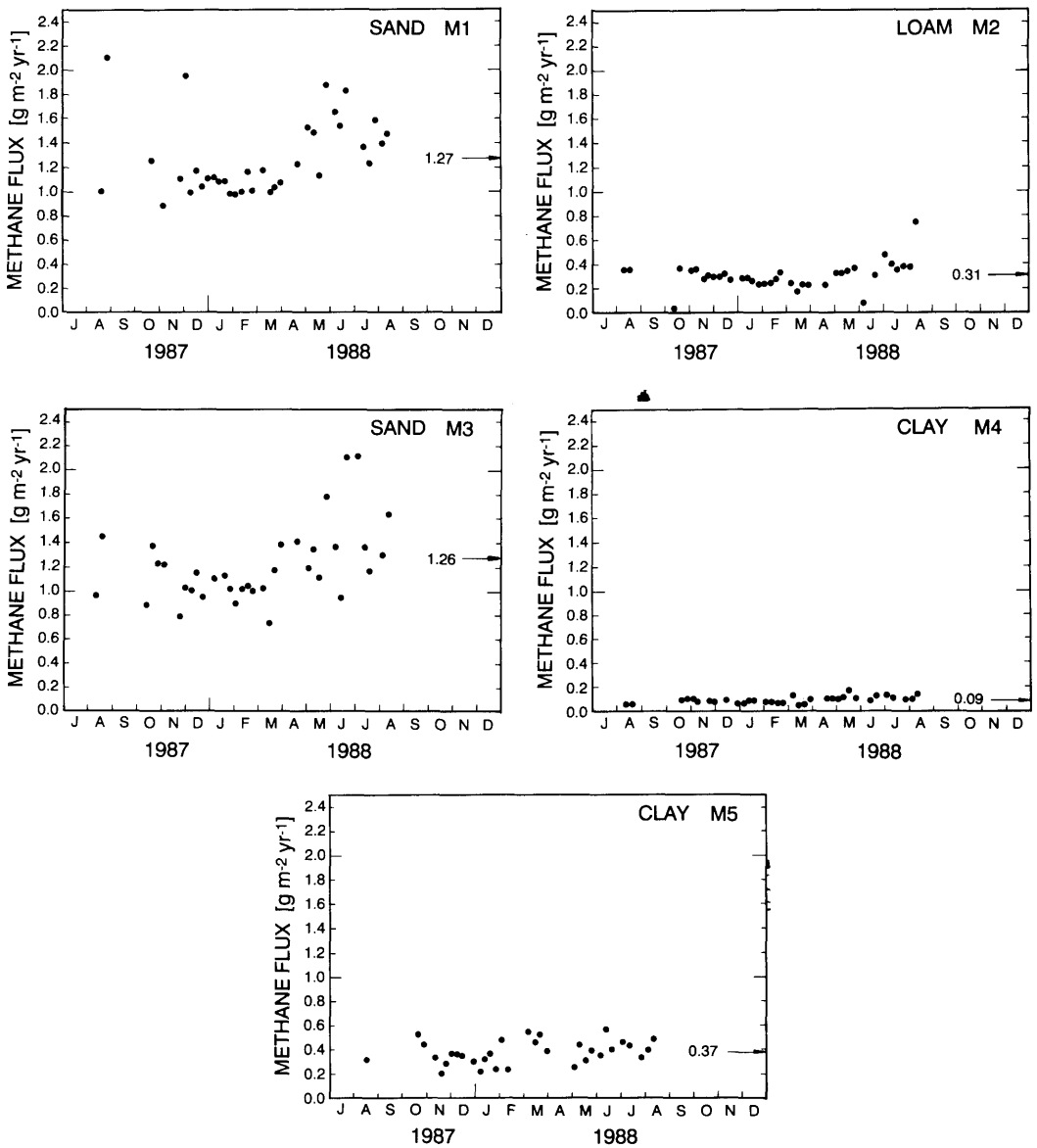

Fig. 2. Seasonal variation of the methane flux from the atmosphere into the soil at 5 different locations in the surroundings of Heidelberg with different types of soil. The methane fluxes have been calculated from the concentration gradient at the soil surface.

zontally (x, y) and exponentially decreasing with depth z (defined as positive downwards):

$$Q(z) = Q_0 \exp(-z/\bar{z}_{CH_4}), \quad (1)$$

integration of the equation of continuity

$$\delta c(z)/\delta t = \operatorname{div} j(z) + Q(z), \quad (2)$$

where $j(z)$ is the methane flux density and $c(z)$ the methane concentration in the soil air, leads to a theoretical steady-state concentration profile which is also decreasing exponentially with depth:

$$c(z) = c_0 \exp(-z/\bar{z}_{CH_4}). \quad (3)$$

The solid lines in Fig. 1 are exponential fits to the measured data points. For this calculation we have used the boundary conditions:

$c(z = 0;\ \text{soil surface}) = c_0 = c_{atmosphere}$,

$c(z \to \infty) = 0$ (more generally, this second boundary condition should read: $\delta c/\delta z(z \to \infty) = 0$, compare Dörr and Münnich, this issue).

Dependent on the nature of the soil the scale height $\bar{z}_{CH_4}$ has been found to be in the range of 9 to 15 cm. The good agreement between measured and calculated profiles confirms the assumptions of horizontal homogeneity and exponential decrease of the methane sink with depth: this distribution of micro-organisms should be expected in a soil with exponentially decreasing soil organic matter.

The methane flux $j(z = 0)$ from the atmosphere into the soil is calculated with Fick's 1st law:

$$j(z = 0) = -P_{CH_4} \times \text{grad}\, c(z = 0), \quad (4)$$

where P_{CH_4} is the permeability of methane in the soil air. This permeability, which depends on the soil parameters porosity, soil moisture, and tortuosity as well as on the molecular diffusion coefficient of methane in air is *a priori* not known. We have calculated the actual permeability at an individual site from the parallely observed ^{222}Radon profile (compare Fig. 1, lower part) and from a direct ^{222}Radon flux measurement at the soil surface. Assuming that the permeability can be expressed as a product of the molecular diffusion coefficient D_0 and a function describing the soil parameters only (Dörr and Münnich, this issue) the methane permeability is:

$$P_{CH_4} = D_0^{CH_4}/D_0^{Rn} \times P_{Rn}$$
$$= D_0^{CH_4}/D_0^{Rn} \times j_{Rn}(z = 0)/\text{grad}\, c_{Rn}(z = 0). \quad (5)$$

Methane fluxes which have been calculated from the concentration measurements of methane and ^{222}Radon in the soil air and from the ^{222}Radon flux at our 5 different sampling sites are plotted in Fig. 2. Surprisingly, the temporal variability of the flux at an individual site is rather low and not more than a factor of two. There are, however, rather large differences in the absolute values at different sites: at sampling sites M1 and M3 (sandy soils) we observe the highest flux values with a mean of 1.3 g CH_4 m^{-2} yr^{-1}. At sampling site M2 (loamy soil) the mean methane flux is only 0.3 g CH_4 m^{-2} yr^{-1}, and at the sites M5 and M4 (clay) mean fluxes of 0.4 and as low as 0.09 g CH_4 m^{-2} yr^{-1} have been observed. The mean methane fluxes show no correlation with soil organic matter content at the individual sites (see Table 1).

As the uptake of methane in the soil has been attributed to microbial decomposition a correlation with soil temperature has to be expected. Fig. 3 shows the correlation of monthly mean values of soil temperature at 15 cm depth and methane flux at site M3. At the same sampling site also the flux of CO_2 produced by root respiration and microbial decomposition of soil organic matter in the upmost 20 cm of the soil has been measured in parallel to the methane flux (Dörr and Münnich, this issue; Dörr and Münnich, 1987). For the CO_2 flux we, indeed, found a strong dependency on soil temperature with a flux ratio $R_{CO_2} = j(15°C)/j(5°C) = 4.3$, whereas the methane flux into the soil is only slightly temperature dependent: $R_{CH_4} = 1.5$.

From Fig. 2, we already concluded that the mean methane flux into the soil varies by more

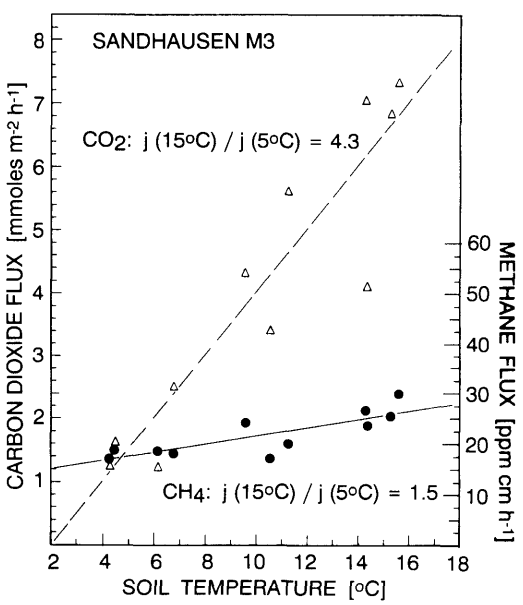

Fig. 3. Correlation between the monthly mean methane flux into the soil respectively the carbon dioxide flux out of the soil and the soil temperature in a sandy soil at Sandhausen (M3).

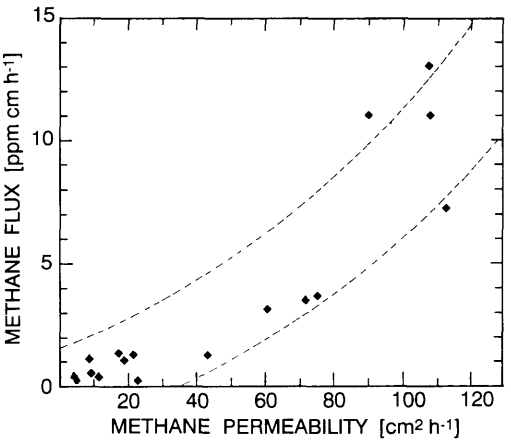

Fig. 4. Relation between methane flux into the soil and permeability of methane in the soil air at different locations in Europe. (The dotted lines are drawn by eye and shall underline the correlation.)

than a factor of ten from one site to another. The individual soil parameters influencing the methane flux from the atmosphere into the soil is the permeability P_{CH_4} of methane in the soil air. Fig. 4 shows the mean methane flux at different sites in Germany plotted against the permeability at each individual site: a high methane flux is only observed at those sites with a large permeability, namely in sandy soils, or, e.g., also in the volcanic soil at Tenerife. The correlation between methane uptake and permeability indicates, that microbial activity is mainly controlled by the gas transport in the soil air rather than by soil temperature. For example, at site M4 no significant summer/winter difference of the methane flux can be observed, but also at sites M1 and M3 with sandy soils the methane consumption rate seems to be controlled completely by gas transport in the soil air. Moreover, the non-linear correlation of the methane consumption with soil permeability (Fig. 4), if significant, indicates that larger methane permeability of the soil might, by some kind of feedback, also increase microbial efficiency.

4. Conclusions

Our measurements of the methane flux into the soil at five different locations in the upper Rhine valley near Heidelberg have shown that the yearly mean methane oxidation rate by soil micro-organisms in aerated soils in temperate forests can be between 0.09 and 1.3 g CH_4 m^{-2} yr^{-1}. Table 2 summarizes the results from this study and observations in other regions, namely in the subtropics and tropics. Under the assumption that our data represent the range of methane flux variation in the investigated areas, the significance of this sink for the global methane budget can be estimated using global area data from Whittaker and Likens (1973) of different cultivation and vegetation types that potentially could act as a net sink of methane. The ranges of the flux are minimum and

Table 2. *Global uptake of methane by different soils*

Vegetation type	Area[a] (10^{12} m^2)	CH_4 flux (g m^{-2} yr^{-1})	Total (Tg yr^{-1})
tropical seasonal forest	7.5	0–0.2[b]	0–1.5
temperate forest	12.0	0.09–1.3[d]	1–15.6
boreal forest	12.0	0.09–1.3[d]	1–15.6
woodland/shrubland	8.0	0.09–1.3[d]	0.6–10.4
savanna	15.0	0.19–0.7[c]	2.9–10.5
temperate grassland	9.0	0.002–0.2[d]	0.02–1.8
cultivated land	14.0	0.002–0.2[d]	0.03–2.8
total			5.6–58.2

[a] Whittaker and Likens (1973).
[b] Keller et al. (1983, 1986).
[c] Seiler et al. (1984).
[d] This study.

maximum values observed in this study and the confidence levels from the mean values given in the literature. From this, the total global consumption rate of atmospheric methane in aerated soils may be as low as 5.6 Tg CH_4 yr^{-1}, and as high as 58.2 Tg CH_4 yr^{-1} corresponding to 1–15% of the total yearly destruction of methane by OH radical, namely about 400 Tg CH_4 yr^{-1} (e.g., Bingemer and Crutzen, 1987). Thus we can conclude that oxidation of methane in aerated soils may have a significant part in the global methane budget. However, further investigation of the temporal and spatial distribution of methane fluxes into the soil in the temperate zone, but also in other climate regions, are needed to obtain representative regional mean values of the methane consumption rate in aerated soils.

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